Attosecond transversal streaking to probe electron dynamics at surfaces

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Attosecond Transversal Streaking to Probe Electron Dynamics at Surfaces

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Abstract The feasibility of attosecond transversal streaking to probe electron transfer dynamics at surfaces and interfaces has been studied. Our simulations suggest that the temporal resolution compares well to existing methods whereas the use of an *s*-polarized streaking field significantly reduces above-threshold photoemission (ATP) and thus also enables the detection of low-energy electrons.

1 Introduction

Electron dynamics at surfaces and interfaces such as photoemission and charge transfer are of fundamental importance in chemical reactions, photovoltaic devices and photocathodes. Recent investigations of the photoemission of diamondoid (*e. g.* tetramantane) coated metals revealed a nearly monochromatic photoelectron spectrum and ultra-short electron transit times.[1, 2] Highly excited electrons from the metal are efficiently transferred to the molecule LUMO, from which they are supposed to be instantly emitted due to the negative electron affinity (NEA).

In a first attosecond condensed matter experiment[3] the relative delay between photoemission of $4f$ and conduction band electrons in W(110) has been determined using the atomic transient recorder (ATR),[4] which is based on longitudinal streaking. Here we propose an experiment to directly determine the electron transit time in diamondoid materials based on transversal streaking. Both the *s*-polarization of the streaking field and the lower intensities that are required lead to a significant reduction of the above-threshold photoemission (ATP) background since ATP electrons are predominantly emitted along the polarization axis.[5, 6]

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2 Simulation

We use classical equations of motion to study the feasibility of transversal streaking for our systems of interest. The parameters used in the simulation correspond to those of the planed experiments. Electrons excited by an XUV pump pulse centered around 40 eV with a pulse duration of 300 as propagate in the NIR field of the probe pulse, which is given by

$$\mathbf{E}_{\mathrm{NIR}}(t) = A_0 \exp\left(\frac{-4\ln 2 t^2}{\mathrm{FWHM}^2}\right)\cos(\omega t + \phi), \tag{1}$$

with a FWHM of 5.5 fs being the pulse duration of the CEP-stabilized driving laser, and ω corresponding to the frequency at 800 nm. Fig. 1 shows how the electron experiences transversal acceleration in the NIR field from the moment of its release, τ, until the pulse passes. The imparted momentum gain is given by

$$\Delta\mathbf{p}(t) = e\int_{\tau}^{\infty}\mathbf{E}_{\mathrm{NIR}}(t)dt = e\mathbf{A}_{\mathrm{NIR}}(t), \tag{2}$$

where $\mathbf{A}_{\mathrm{NIR}}(t)$ is the vector potential in the Coulomb gauge. The change of the parallel momentum, $\mathbf{p}_{\parallel}$, leads to a change of the emission angle, θ, which can be detected as a function of time-delay between XUV and NIR pulses by an angle-resolved electron analyzer. Initial momentum distributions were taken from experimental data, which was obtained by angle-resolved photoemission spectroscopy (ARPES) using He II$_\alpha$ (40.8 eV) radiation.

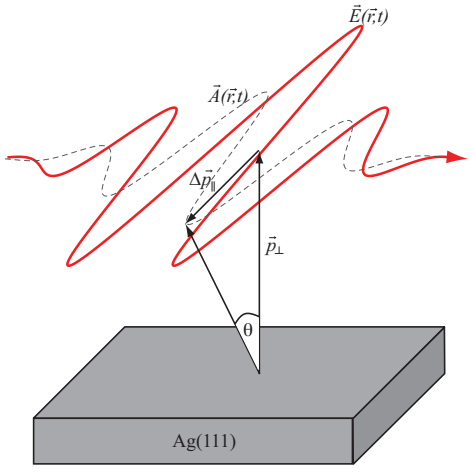

Fig. 1 Principle of transversal streaking. An electron emitted normal to the Ag(111) surface with initial momentum, $\mathbf{p}_{\perp}$, experiences transversal acceleration in the *s*-polarized NIR field, $\mathbf{E}(\mathbf{r},t)$, leading to a change of the parallel momentum, $\mathbf{p}_{\parallel}$ proportional to the vector potential, $\mathbf{A}(\mathbf{r},t)$, and thus to a net-change in emission angle, θ.

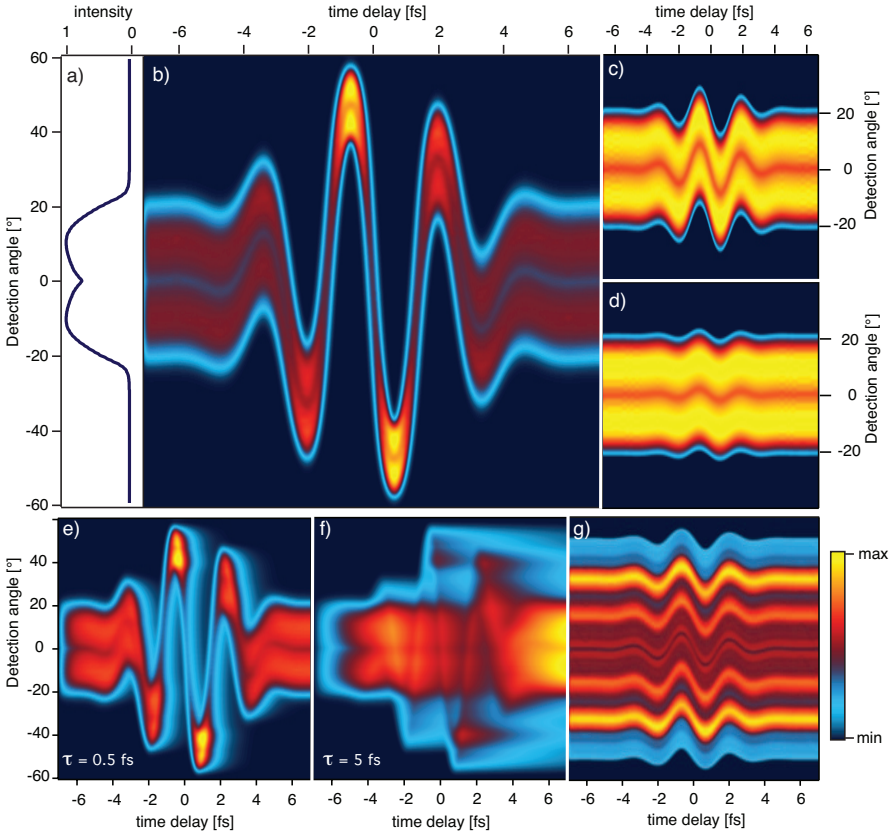

Fig. 2 (a) Initial momentum distribution of NEA electrons in the laboratory frame. (b-d) Simulated streaking spectrograms for given initial momentum at field intensities of $I_{\mathrm{NIR}} = 5 \times 10^{12}$, 1×10^{11} and 5×10^{9} W/cm^2, respectively. (e,f) Spectrograms for non-instantaneous emission described by exponential decay functions with indicated time constants. (g) Spectrogram of Ag(111) 4*d* valence band electrons at $I_{\mathrm{NIR}} = 5 \times 10^{12}$ W/cm^2.

3 Discussion

The initial momentum distribution of the NEA electrons that was used in the streaking simulations is shown in Fig. 2a. The spectrograms that were obtained for this distribution at different streaking field intensities are presented in panels 2b-d. The slow NEA electrons (E_{kin} = 0.5 eV) still experience a deflection that is large enough to be detected in an angle-resolved experiment at field intensities as low as 5×10^{9} W/cm^2. In a case where electrons are not emitted instantaneously from the tetramantane LUMO we simulated the spectrograms taking an exponential decay into account. The spectrograms we obtained for time constants of 0.5 and 5 fs are given in panels 2e and 2f, respectively. Interestingly, even for a lifetime nearly twice as long as one optical cycle of the streaking field, the shape of the vector potential,

$\mathbf{A}_{\mathrm{NIR}}(t)$, and therefore temporal information can still be retrieved. Finally, the spectrogram of an initial momentum distribution that was obtained by a constant-energy-cut through the Ag(111) 4*d* band is shown in Fig. 2g. Those electrons serve as reference to clock the LUMO transit time since they were directly emitted from the Ag(111) valence band. The same field that is used to streak the slow NEA electrons is strong enough to deflect the much faster 4*d* electrons (E_{kin} = 36 eV).

Some preliminary experiments concerning the polarization-dependence of above-threshold photoemisson (ATP) that we performed at low field intensities on the order of 10^9 W/cm^2 on pristine and tetramantane-covered Ag(111) indicate that in particular the high-energy tail of the ATP spectrum is significantly suppressed for *s*-polarized light.

4 Conclusion

Our simulations suggest that transversal streaking is a valuable alternative over longitudinal streaking using the atomic transient recorder (ATR) technique, which offers two distinctive advantages. (1) The *s*-polarized streaking field leads to a significant reduction of ATP backround. (2) Data acquisition time can be reduced using 2D detectors, which simultaneously use the energy and angular resolution of a hemispherical electron analyzer. This is important for surface experiments since many well-prepared systems have a very limited longterm stability.

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